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FALLOUT

Radioactive fallout represents nuclear debris deposited on the ground from the atmosphere. Although most fallout originates from tests of nuclear weapons, some may also arise from specific nuclear operations. Depending on the method of deposition, fallout is characterized as dustout--deposition of dry solids; rainout--carried by rainfall; or snowout--deposited by snowfall.

When nuclear weapons are detonated, the fission products formed are carried aloft. The altitude attained by the fission fragments is governed by the nature of the detonation, whether a high altitude shot, a tower shot, subsurface, or underwater shot; and the energy equivalent of the shot, whether in the kiloton or megaton range. The area of fallout is governed by the above factors as well as the meteorological pattern at the time of the detonation, the location of the test site, etc.

When a nuclear device is exploded, the products formed include the fission products (elements found in the middle of the periodic table generally ranging from germanium to gadolinium), unfissioned fuel (uranium, plutonium), and induced radioactive products (from the metal components of the weapon, and from neutron activation of environmental materials like calcium, phosphorus, sulfur, etc).

The altitude at which the detonation takes place will govern in part the immediate fallout area. For example, in the case of a high

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* See page 14 for footnotes

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altitude shot, the fission products will be carried upward by the fireball and the altitude attained will be a function of the energy equivalent of the shot. As a result, the fission products will not be deposited near the site of the detonation but at some distance away depending on the meteorological pattern. In the case of a subsurface or underwater shot, the fallout would be associated with soil or water nuclei and would be deposited much closer to the site of detonation. Fallout would be much more localized in this instance.

Those radionuclides which have short half lives will decay out and will not reach the ground in the case of a high altitude shot involving megatons of energy, but the longer-lived radionuclides will be carried away from the test site and deposited over a much larger area for a longer period of time. Shorter-lived radionuclides will be present in localized fallout from low altitudes, including subsurface or underwater detonations, or low energy shots. In this connection, it may be pointed out that ratios of specific radionuclides may be of value in estimating the age of fallout. For instance, ratios of Sr^{89}: Sr^{90}, Ru^{103}: Ru^{106}, or Ce^{141}: Ce^{144} may be used for this purpose.

Information published by Machta (1) and Martell (2) has shown that approximately twice as much fallout has been deposited in the northern hemisphere as has been deposited in the southern hemisphere, with major latitudinal stratification in the 40 to 60° North latitude band. This is due primarily to a discontinuty in the tropopause, the zone which separates the troposphere from the stratosphere. Fallout

leaks into the troposphere through this discontinuity, accounting for the greater degree of fallout in this area. A similar discontinuity exists in the southern hemisphere. Stratospheric fallout originally was estimated to have a residence time of approximately 7 to 10 years, which would have permitted substantial decay of most fission products with a half life of one year or less, but more recent studies (2) show that the residence time may be less than one year. Thus, there is a greater areal distribution of younger material, whereas, in the past, short-lived materials were associated primarily with localized fallout. This more rapid return of fission products from the stratosphere means that higher concentrations of radioactive material will be deposited sooner, increasing the levels of contamination of our water and food sources.

As stated earlier, fallout will also occur from nuclear industrial operations and will be released during normal operation of reactor facilities and spent nuclear fuel chemical reprocessing plants. These discharges will be controlled and should not add materially to the amount of nuclear debris contributed to a given area. However, during an accident or nuclear incident, such as the one which occurred at Windscale, England, sizeable increases in activity may occur.

WATER ENVIRONMENT

Fallout, which is carried down mainly by rainfall, contaminates our water resources and agricultural areas. The amount of this material actually reaching the soil will be governed by the nature and

depth of vegetative cover and upon the intensity and duration of precipitation. Radioactive debris reaching the earth will be either in solution or associated with particulate matter. Shortly after a detonation, the major fraction of the radioactivity found in rainfall will be associated with the suspended fraction. As residence time increases (older material reaching the ground), more material is in solution.

Radioactive debris carried by rainfall will be intercepted by plant surfaces and, depending on the nature of the plant surface, and intensity and duration of precipitation, variable amounts of radioactivity will be deposited on such surfaces. Rainfall may also fall upon the soil and remain in contact with it in accordance with the runoff characteristics of the given area. Runoff will be most rapid from hilly terrain and impervious type soils, and will be reduced in flat, open country. The slower the rate of runoff the greater the opportunity for decay and removal of the radioactive materials contained in the rainfall. Particulate matter will be intercepted by filtration and soluble radionuclides may be absorbed, adsorbed, or exchanged on contact with soil particles. Silty and clayey soils have higher exchange capacities than do sandy or graveley soils. Soils high in organic matter will also remove radioactive materials by any of the processes indicated above. Residual cationic, anionic, or non-ionic radionuclides will be carried off with runoff and will eventually find their way to surface waters either through surface runoff, as just described, or via the ground water route. In addition to this residual radioactivity, surface

waters will contain those radionuclides deposited directly into the water itself.

Further removal of radioactivity takes place in the water environment. There is the loss of radioactivity due to decay of the shorter lived radionuclides. Biological life--plankton, algae, benthic forms, fish, etc.--in the water may remove specific radionuclides through metabolism, absorption, adsorption, or exchange. Dead or decaying organic matter present will also remove radioactive materials as will bottom silts or muds. In addition, activity adsorbed or exchanged on turbidity may settle out with a diminution of stream velocity. In all cases, it must be recognized that removal as such is a misnomer. A holdup takes place by the transfer of activity from the liquid medium to those solid phases associated with the liquid phase. The holdup is temporary, because radionuclides taken up by biological material may be released to the liquid phase upon death or destruction of the biological forms involved. Similarly, radioactivity associated with bottom muds and silts may be moved and redeposited at some downstream location during periods of flood resulting in a higher level of contamination at these points, as well as subjecting agricultural areas to additional fallout contamination from overflowing streams or rivers.

Potable Water

Streams or rivers may be used as sources of potable and industrial water supply, irrigation water, recreation, fishing, or navigation. Each of these uses is influenced by the quality of the water. Contamination

by fallout must be considered in determining water quality along with turbidity, hardness, or any other chemical, bacteriological, or biological contaminant.

Fallout present in streams used as sources of potable water will be removed in part during treatment of the water (3, 4, 5). To obtain some insight as to the amount of removal that can be expected, stream waters are analyzed for gross activity associated with particulate matter as well as that in solution. Radioactivity associated with particulate matter such as silt, biological materials, etc., will be removed essentially with the removal of the silt or biological material by coagulation, sedimentation, and filtration. Higher gross removals may be expected by water treatment plants when relatively fresh fission products are treated and when the major fraction of radioactivity is associated with particulate matter. As the material becomes older, i.e., contains more longer-lived radionuclides, removal efficiencies may be lowered, particularly where routine coagulation and filtration are employed. Treatment involving lime and soda ash softening or ion exchange will be more effective for the removal of some of the longer-lived materials, as for example, strontium.

Streams are not the only sources of surface water supply. Ponds or lakes must also be considered. In general, these should have higher activity levels, inasmuch as their activity is retained and accumulated following successive periods of rainfall in contrast to river waters

which pass the material on downstream immediately. Cisterns serve as another source of water supply. Levels of activity in cisterns are similar to levels of radioactivity encountered in rainfall.

Industrial Water

Industrial waters used for food processing and preparation should be low in the more hazardous radionuclides--strontium and cesium. In the production of paper or photographic chemicals and film, waters of low activity are necessary. Waters contaminated with radioactivity used for the production of certain chemicals may have to be extensively treated to avoid contamination of reagent grade chemicals.

Irrigation

Where water is used for irrigation, there will be uptake by plants from the water as well as from the soil. In spray irrigation, the aerial portions of the plant will be contaminated and there will be an increased level of fission products in the upper soil layers due to absorption and exchange. This might eventually result in a higher translocation rate from soil to roots to leaves. Furthermore, laboratory data show a higher rate of uptake in plants grown in liquid nutrients (hydroponic studies) than similar plants grown in soils contaminated at the same level.

Other Uses

Radioactivity in bottom muds and silt will contaminate boats, fishing gear, and other sports equipment, and may serve as an increased hazard to bottom feeding fish. Similarly, fish who feed primarily on biological forms may be affected by the uptake of activity by such organisms.

Seasonal variation in uptake has also been demonstrated. During summer months, when there is an increase in plankton and other forms and when higher metabolic rates prevail in fish, higher levels of activity have been found in fish (6, 7).

Levels of Activity

Studies carried out by the Public Health Service as a part of its National Water Quality Network, which includes 55 stations on 16 major rivers and the Great Lakes, have shown that gross beta-gamma activity values have averaged 56 μμc/l, of which about 48 per cent was dissolved. Gross alpha activity levels averaged 3.7 μμc/l (8). More recently strontium-90 determinations have been made on two series of 13 week composites of these samples and have shown levels ranging from 0 to 4.8 μμc/l with a geometric average of 0.9 μμc/l for the period January-March 1959 and 1.4 μμc/l for the period April- June 1959. For the two stations we have in Texas the gross dissolved activity level reached a maximum value of 63 μμc/l, and the Sr^{90} 1.1 μμc/l (9).

SOIL ENVIRONMENT

The soil investigation program of the AEC has been summarized by Dr. Lyle Alexander of the Department of Agriculture and the effect of strontium-90 on soils has been reported as follows (10):

"Most Sr^{90} reaches the surface of the earth in the water brought down from air by rain or snow. To a lesser extent, the formation of dew on vegetation and the impingement of fog on plants make a contribution to the total amount reaching the soil. The almost complete absence

of this fission product (Sr^{90}) in the west coast of Chile, where rainfall in essentially zero, is evidence that very little strontium-90 falls out without moisture as a carrier. These statements, of course, do not apply to coarse grained materials that may fall out close to test sites. They do apply to materials that have moved hundreds of miles either in the troposphere or the stratosphere.

"Most soils retain strontium-90 against leaching by rain water in the same way that they retain the plant nutrient element calcium. There is gradual movement downward in proportion to the amount of water passing through the soil into the ground water. The process is very slow and even in areas of high rainfall it takes several years to move strontium-90 downward a few inches. Because of this retention, properly selected soils can give a good integrated value of the strontium-90 that has fallen to the ground in a selected location.

"However, all soils do not stay in place during a rainstorm and many soils that do stay in place do not take in water as fast as it falls. Hence, the strontium-90 that falls evenly on a given acre of soil is non-uniformly distributed after the movement of the rain water and soil has ceased. A well grassed or wooded area that is level and receives no water from higher lying ground should retain essentially all of the strontium that falls on it. A steep or stony area that absorbs little of the rainfall would retain little strontium-90.

"Areas at the foot of slopes and in stream bottomlands that receive soil from higher lying ground by alluvial or colluvial action will have more strontium-90 than fell upon them in the snow and rain.

"There is a special problem in regions such as our Plains States where the winter snows drift into fence rows and hedges and into gulleys. A smooth unprotected area loses some of the strontium-90 falling on it, while places where snow accumulates get more strontium-90 than reached them by direct deposition in snow. In such localities there are few, if any, sites that can be said to have the average amount for the area as a whole."

Plant Contamination

Vegetation may become contaminated by three different mechanisms:

1) superficial contamination due to airborne debris lodged on leaves and stems;

2) internal contamination due to material absorbed from 1); and

3) internal contamination due to absorption through roots and subsequent translocation.

Immediately after a detonation, superficial contamination alone would be expected. With time, as additional radioactivity was deposited on and carried into the soil, there would be the translocation of radioactivity from the soil through the root systems to the internal portions or by material absorbed directly into the leaves and stems from that deposited thereon. In many samples of vegetation that have been analyzed, higher levels of radioactivity have been observed in the old leaves with low level activity in the roots which suggests that absorption through leaves is important. Grasses collected in the field after the Hurricane shots at Monte Bello (Australia) showed little preferential

accumulation of strontium relative to ruthenium or cesium. However in plants grown under experimental conditions in which fission products could only be absorbed through the roots there was a marked preferential absorption of strontium. Also in support of absorption through the leaves rather than through the roots was the observation that the bulk of the activity in the soil was retained in the upper inch, which was above the level of the majority of the roots and was furthermore so dry that little absorption could be expected (11).

From the above statements it is quite obvious that solubility is an important factor in evaluating biological availability of the fission products. Availability is also characterized by determining the water soluble, exchangeable, and non-exchangeable fractions in the soil (12). Movement of ions through the soil is greatly affected by the pH of the soil. More of the ions would be released in an acid soil than from neutral or alkaline soils, for example.

Generally speaking, different soils react differently in their ability to make available to root systems the radionuclides contained therein. Specie differences as far as plants are concerned have also been observed (13). The uptake of strontium-90 could be reduced by increasing the calcium content of soils deficient in this element. Where sufficient calcium was present in the soil the addition of more calcium had no effect on Sr^{90} uptake. Addition of stable strontium caused a slight increase in the uptake of Sr^{90}. Sr^{90} uptake was decreased in the presence of soil organic matter. This was attributed to greater biological activity and binding of strontium in biological forms making the

Sr^{90} less available to roots (14).

The suggestion has been made of utilizing plants themselves for decontaminating soil. But studies (12) show cropping to be a very slow process as evidenced by the following data: The highest percentages of fission products removed by cropping were: Sr^{90} 8.68%; Ru^{106} 0.13%; Cs^{137} 0.48%; Ce^{144} 0.02%.

To measure the influence of cover crops in reducing the amount of fallout reaching the soil surfaces, the radioactivity of soil supporting a good yield of red clover and wheat at varying distances from ground zero were compared. At 7 mi from ground zero wheat and red clover retained 0.1 and 0.2% of the total fallout per unit area of soil, at 48 mi one to 5%; and at a distance of 106 mi, as much as 25% of the total fallout was retained on vegetation. Increasing retention with distance appeared to be a function of particle size (12).

The amount of fallout particles retained by plant foliage is dependent on such characteristics as hairs, glands, stomata, and other mechanical traps. The major fraction of fallout particles retained on foliage had a diameter of less than 44 microns with an average size of approximately 20 microns. Fallout particle retention by plants was also investigated by immersing plant material in distilled water, in 0.1 N HCl, and 5 per cent Versene solutions. Distilled water removed from 36 to 97 per cent of the fallout activity, 0.1 N HCl 48 to 98 per cent, and Versene 54-98 per cent. Radioactivity was readily removed from smooth, flat, waxy leaf surfaces by each solution (12).

Much of our exposure to strontium-90 will be through the diet. Measurements we have made on various foods show great fluctuation in levels depending on the geographical area in which they were grown as well as climatological conditions during growth (8, 15). Studies carried out by Middleton in England (16) using foliar spraying showed that less than 0.1 per cent of the Sr^{89} initially deposited on the plant was found in the grains from ears which emerged after spraying. When plants were protected from rain after spraying, the strontium-89 content of the grain was significantly lower than when the plants were exposed to rain. The strontium-89 content of the grain was more than 20 times greater when spraying occurred after the ears had emerged but while the grain was still immature, and the values for the final spraying just before harvest were only slightly lower. From this he concluded that "It is therefore possible that the period shortly after the ear emerges is critical with respect to the contamination of wheat grain by strontium-89."

From the above discussions it can be seen that retention of fallout on soil and agricultural products depends upon a great many factors not all of which have been defined as yet, or are easily definable.

Considerable information exists on the environmental levels of radioactivity, particularly from the standpoint of soil deposition, plant uptake, and levels in other media, but little information is available as to the significance of these levels in terms of man's

exposure. To obtain answers to this latter question will require the combined talents of many individuals representing a multiplicity of professional disciplines.

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